LA-UR-15-29203
Approved for public release; distribution is unlimited.

Title:	Multiplicity Counting
Author(s):	Geist, William H.
Intended for:	Presentation for programmatic work
Issued:	2015-12-01

Multiplicity Counting

MPF

Neutron Counting – Background and Review

Attributes of Neutron Counting

- Neutrons are highly penetrating
  - Neutron signatures are sometimes the only way to rapidly assay large/dense samples.
- Neutron counting data can be collected quickly
  - Materials of interest tend to be prolific neutron-producers
  - Nominal sample prep
- Spontaneous Fission Neutrons
  - The primary signature of the even isotopes of plutonium.
- Induced Fission Neutrons
  - A signature for both fissile plutonium and uranium.
- Need isotopic information to get **total** mass.

Neutron Counting Quantities

When performing a neutron assay measurement we generally have three unknown quantities that are specific to the sample:

m = the mass of material ($^{240}Pu_{eff}$, *more on this later*)
M = the sample leakage multiplication (*more on this too*), and
α = the ratio of (α,n) neutrons to spontaneous fission neutrons produced in a sample (*we'll talk about this too*)

We also have known detector-specific quantities that are provided in the documentation and/or the software configuration provided by SGTS:

ε = the detector efficiency
τ = the neutron die-away time of the detector
δ = detector dead-time correction coefficients (A, B, C, δ_{mult})
G, P_d = Coincidence gate and pre-delay settings

History of Neutron Counting

- **Gross Neutron Counting (Counts, Totals, Singles)**
  - The first neutron assay instruments used the gross neutron signal to deduce assay information.
  - Accurate assays could be obtained for only limited material types.
  - Yields one observable value (Counts)
- **Neutron Coincidence Counting (Reals, Doubles)**
  - "Coincidences" are time-correlated pairs of neutron counts.
  - Two kinds: Reals and Accidentals
  - Offers a greatly expanded set of applicable materials for accurate assay.
  - This technique has wide application for international safeguards.
  - Large errors can occur in the assay of impure materials.
  - Yields two observable values (Totals, Reals).
- **Neutron Multiplicity Counting (Triples)**
  - An extension of neutron coincidence counting; Moments Analysis
  - Adds counting of time-correlated triplets of neutron counts
  - Can dramatically improve neutron assay accuracy.
  - Yields three observable values (Singles, Doubles, Triples).

Terminology

With the evolution of counting techniques, came an evolution of terminology of the measured values:

	Gross Counting:	Coincidence Counting:	Multiplicity Counting:
Signal:	1 Counts	1 Totals 2 Reals	1 Singles 2 Doubles 3 Triples
Background:	1 Background	1 Background, Totals Background 2 Accidentals	1 Background, Totals Background 2 Accidentals, Doubles Background 3 Background Distribution, Triples background

Terminology

The term, *multiplicity*, is used in many ways:

- A neutron coincidence counter or "neutron *multiplicity* counter".
- The technique of "*multiplicity* counting" based on the use of special electronics to get higher order statistical information from a shift register circuit.
- Nuclear data of the fission process -- i.e. the emitted "multiplicity distribution of fission events".
- The statistical information itself -- i.e. the "*multiplicity* of events" in a coincidence gate.
- The theoretical "*Multiplicity* Point Model" used to reduce the measured information to an assay result.

Application of Multiplicity Counting

Multiplicity counting has advantages over coincidence counting:

- Calibration does not require a representative set of standards.
- Accuracy of between 1 to 3% can be obtained for most material types.
- Appropriate counters attain high precision.
- Technique can be used on many material types:
  - Pure
  - Impure
  - Material with out standards
  - Oxidized Pu metal
  - Metal or oxide
  - Scrap and waste

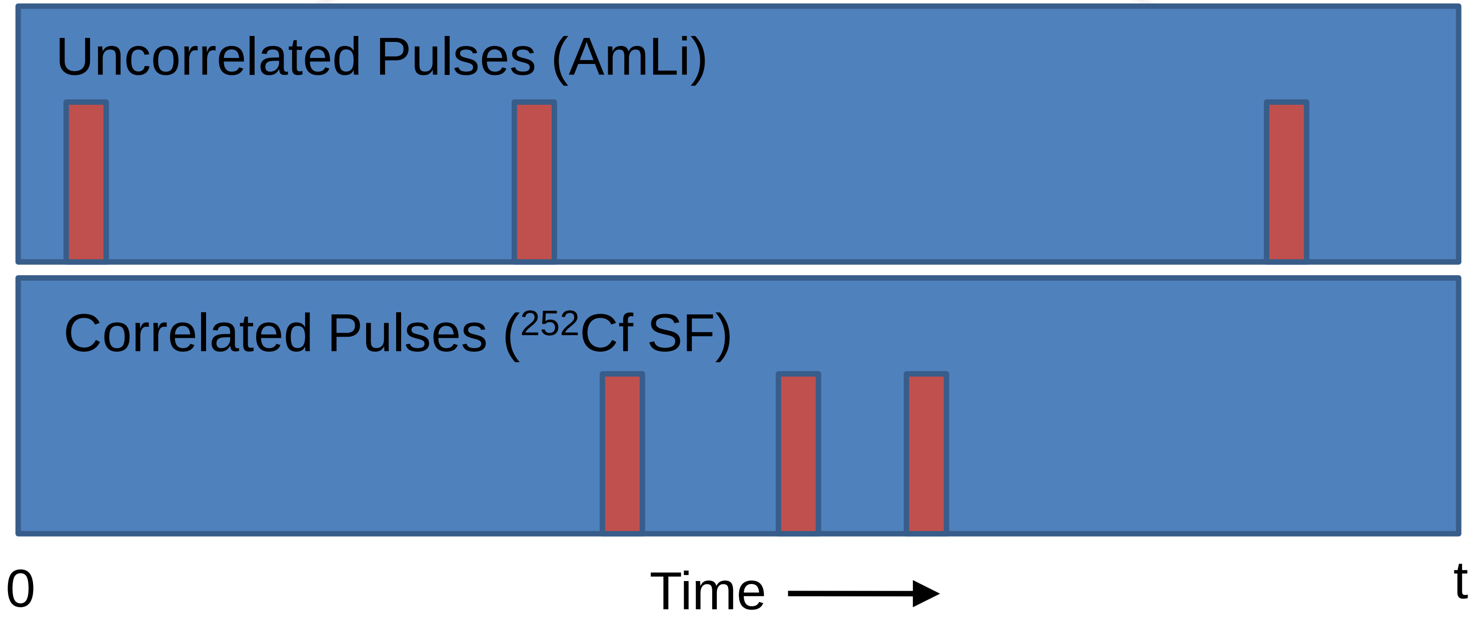
Neutron Signatures

Mechanisms behind the production of neutrons:

- All neutron signatures are **statistical** in nature.
- A **fission event** is a **random** process. If more than one neutron is emitted, those neutrons are **coincident in time**.
- Neutrons produced from the interaction of an **alpha particle** with a light element nucleus are always emitted **singly**. This is also a **random** process.

One cannot distinguish neutrons from different processes except by statistical methods.

Concept: Correlated Pulses



Two pulse trains over the same time interval.

Both have the same count rate.

- Pulses from a single fission are, on average, closer together in time than pulses from different events.
- We want a device to distinguish between these two distributions.

Properties of Neutrons

- Highly Penetrating
  - Do not strongly interact with matter; zero charge
  - Not *easily* shielded

    Exceptions:

      Hydrogenous materials (water, polyethylene, etc.)

      Neutron poisons (Cd, Gd)

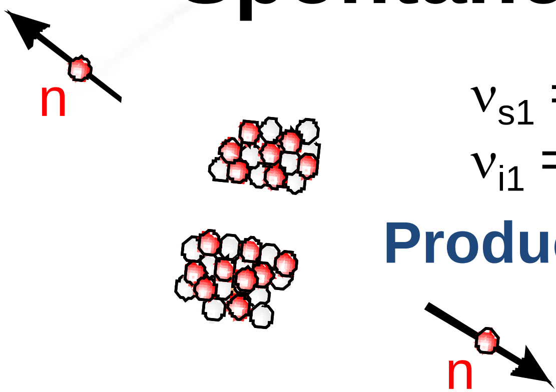
  Consequence:

    One can usually measure entire volume of item.

- Produced by 3 processes.
  - Spontaneous fission
  - Induced fission
  - alpha-n reactions
- **Fission** is the signature that is useful for assay.

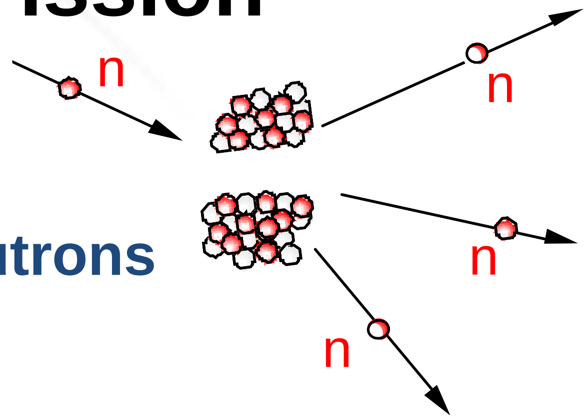
Sources of Neutrons

Spontaneous & Induced Fission

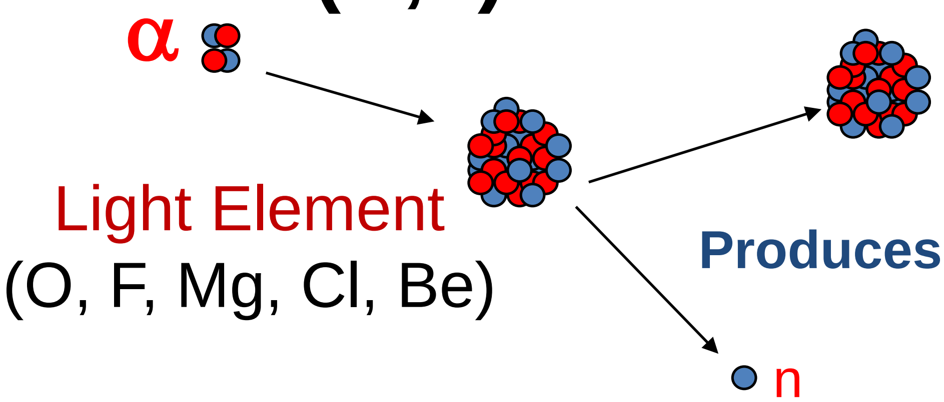


ν_{s1} = 2.154 (for ^{240}Pu)
ν_{i1} = 3.163 (for ^{239}Pu at 2 MeV)

Produces a multiplicity of neutrons



(α,n) Reactions



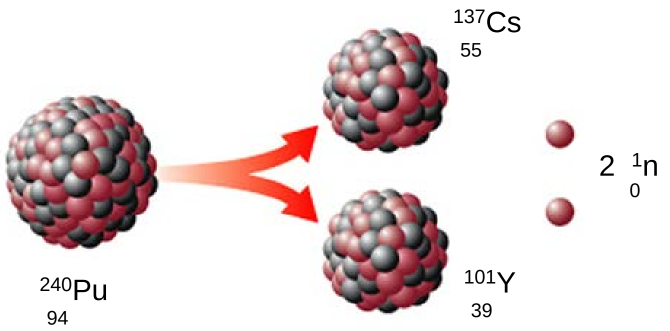
Produces <u>only</u> single neutrons

Spontaneous Fission

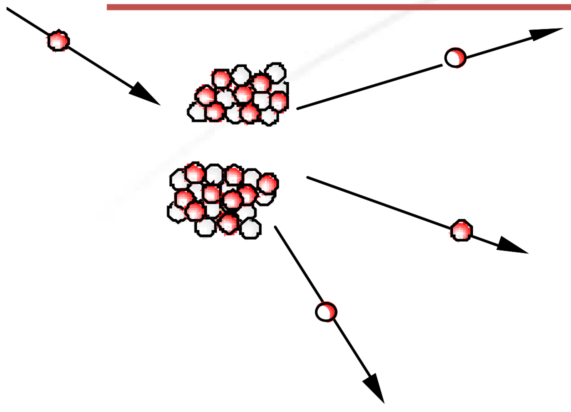
Important Facts:

1. **Emission is in bursts from 0 to 6 (or more) neutrons.**
2. **Even-numbered isotopes tend to decay by spontaneous fission (fertile isotopes)**
3. **Emission Rate is independent of chemical composition.**

Nuclide	Production rate (n/s/g)
^{238}Pu	**2500**
^{239}Pu	0.022
^{240}Pu	**1020**
^{241}Pu	0.024
^{242}Pu	**1730**
^{252}Cf	2.3×10^{12}
^{244}Cm	1.08×10^{7}



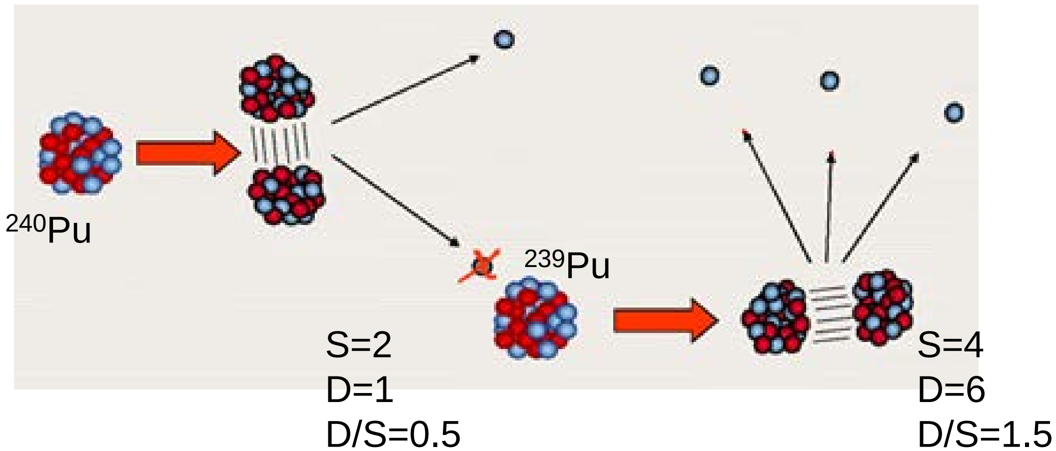
Induced Fission & Multiplication



ν_{i1} = 3.163 For ^{239}Pu at 2 MeV

- A neutron present a sample can either induce a fission, escape from the sample or be captured in the sample

- Multiplication occurs when fissions are induced in a sample by neutrons born in the sample

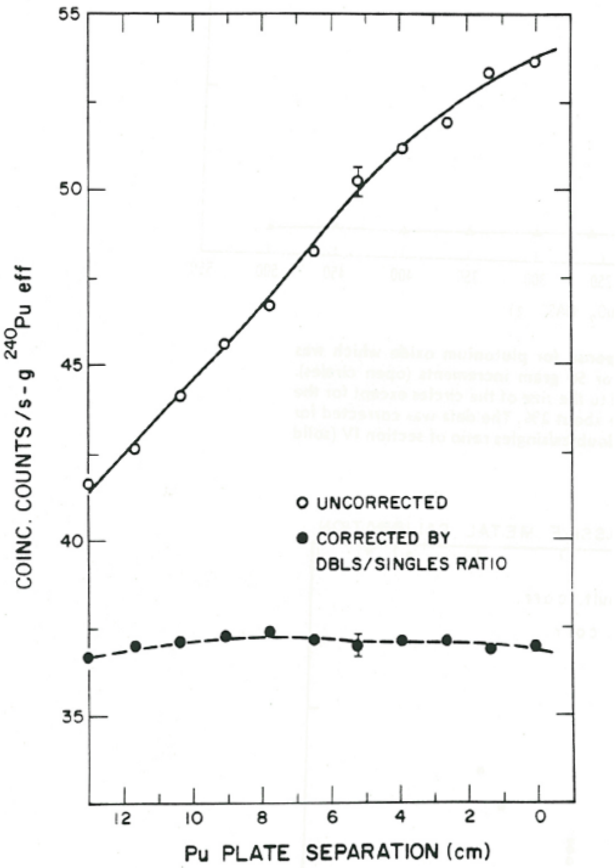


Where we had 2 neutrons, we now have 4.

We know the “4” resulted from multiplication because the D/S ratio changed

Induced Fission & Multiplication

Two 1-kg Pu plates moved closer together



We observe that the doubles rate is changing with the sample geometry.

We can correct for this using the change in D/S ratio to arrive at the desired case where the corrected doubles rate is no longer a function of geometry.

This is the basis of the "Known-Alpha" analysis (aka "multiplication-corrected reals")

The induced fission probability, p, in a sample depends:

mass, geometry, density, isotopic- and chemical-composition

Induced Fission & Multiplication

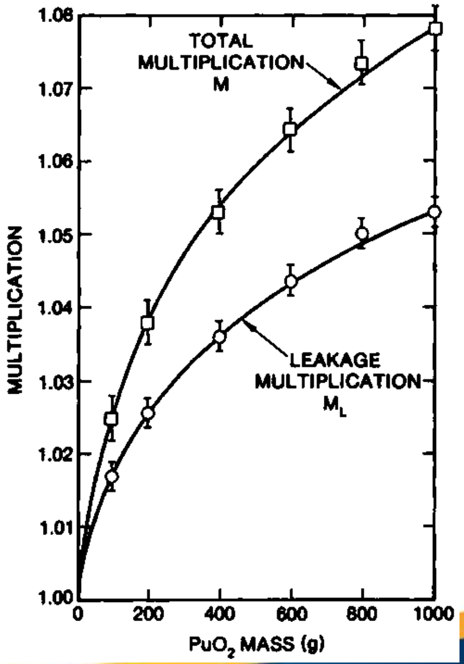
3 things can happen to a neutron in a sample

$$p + p_L + p_C = 1$$

p = probability that a neutron will induce a fission in the sample
p_L = probability that a neutron will escape the sample (leakage)
p_C = probability that a neutron will be captured in the sample (assumed to be small)

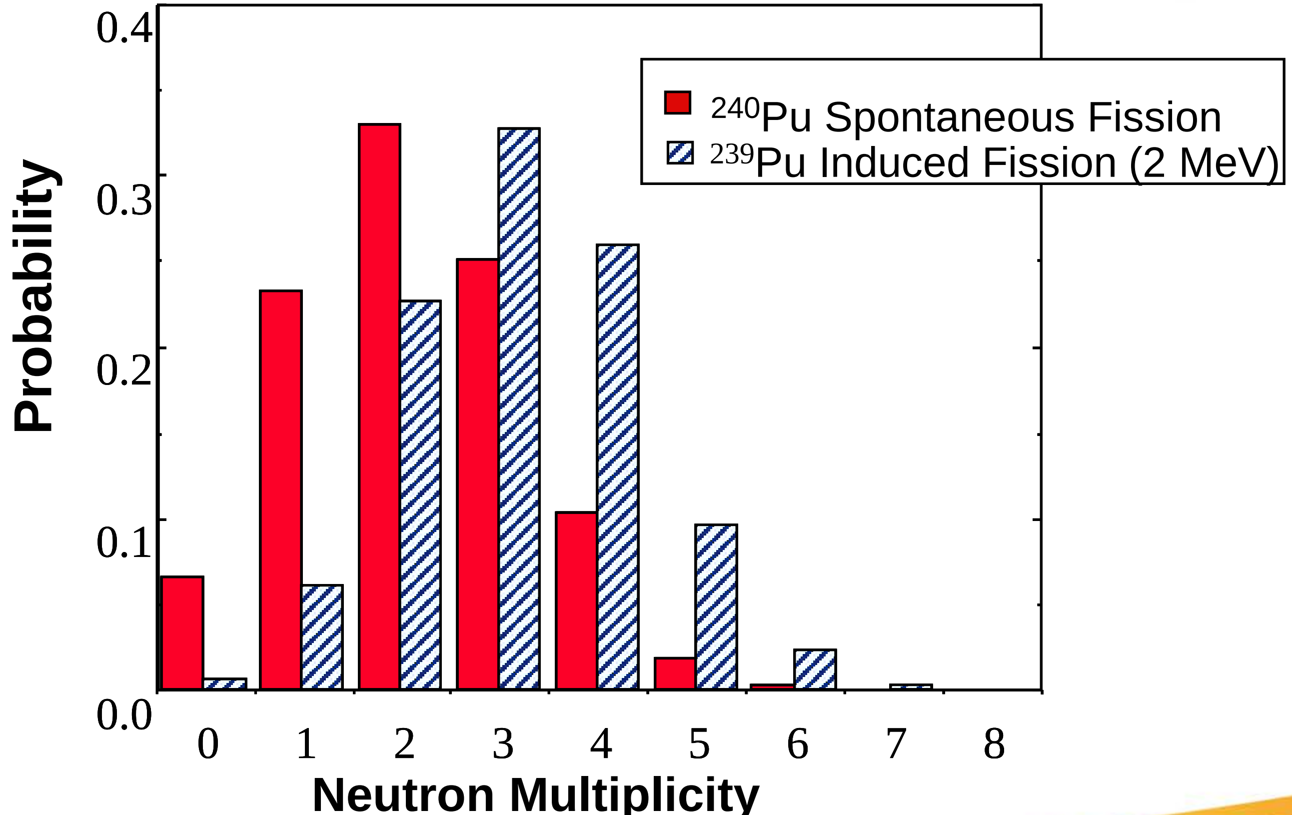
In neutron counting we are concerned with Leakage Multiplication, M_L:

$$\mathbf{M_L = M_{Tot} p_L = (1-p)/(1-p\nu_{i1})}$$



Fission Neutron Multiplicity Distributions

Why do we keep saying 2.15 and 3.16 neutrons are produced per fission?



(Alpha, n) Reactions

Important Facts:

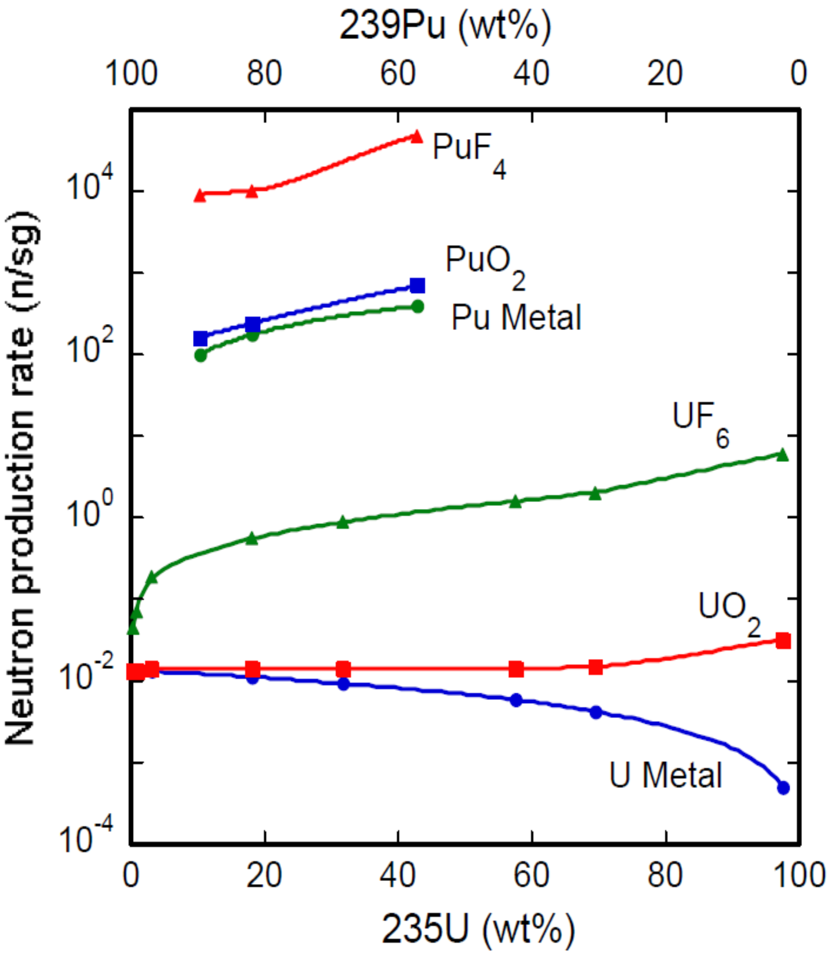
- Uranium and Plutonium both emit alpha particles that can react with the nuclei of light element matrix components to produce more neutrons.
- Actinides tend to decay by alpha-decay
- Americium-241 is an important prolific alpha emitter in Pu (from the β-decay of Pu-241)
- (α,n) neutrons are emitted randomly and singly.
- Neutron emission is dependent on the chemical composition of the sample.

Example Materials: AmLi, PuO_2, UF_6

We Define:

α = ratio of (α,n) neutron production to spontaneous fission neutron production.

Values of α



Pu metal: α = 0
U metal: α = 0

PuO_2 (reactor grade, 57% Pu-239)
α = 0.84
PuO_2 (weapons grade, 93% Pu-239)
α = 0.78

PuF_4 (reactor grade, 57% Pu-239)
α = 135
PuF_4 (weapons grade, 93% Pu-239)
α = 110

$^{240}Pu_{eff}$ mass

$$^{240}Pu_{eff} = 2.52\ ^{238}Pu + {}^{240}Pu + 1.68\ ^{242}Pu$$

The $^{240}Pu_{eff}$ mass is the mass of pure ^{240}Pu required to produce the same neutron doubles count rate as that produced by the mix of fertile isotopes in the actual sample.

Neutron analysis determines the $^{240}Pu_{eff}$ mass.

To determine the total mass from $^{240}Pu_{eff}$, the isotopic values are needed.

A Quick Review

We have discussed three attributes of a verification item that we need to sort out:

$^{240}Pu_{eff}$ mass – Our verification objective

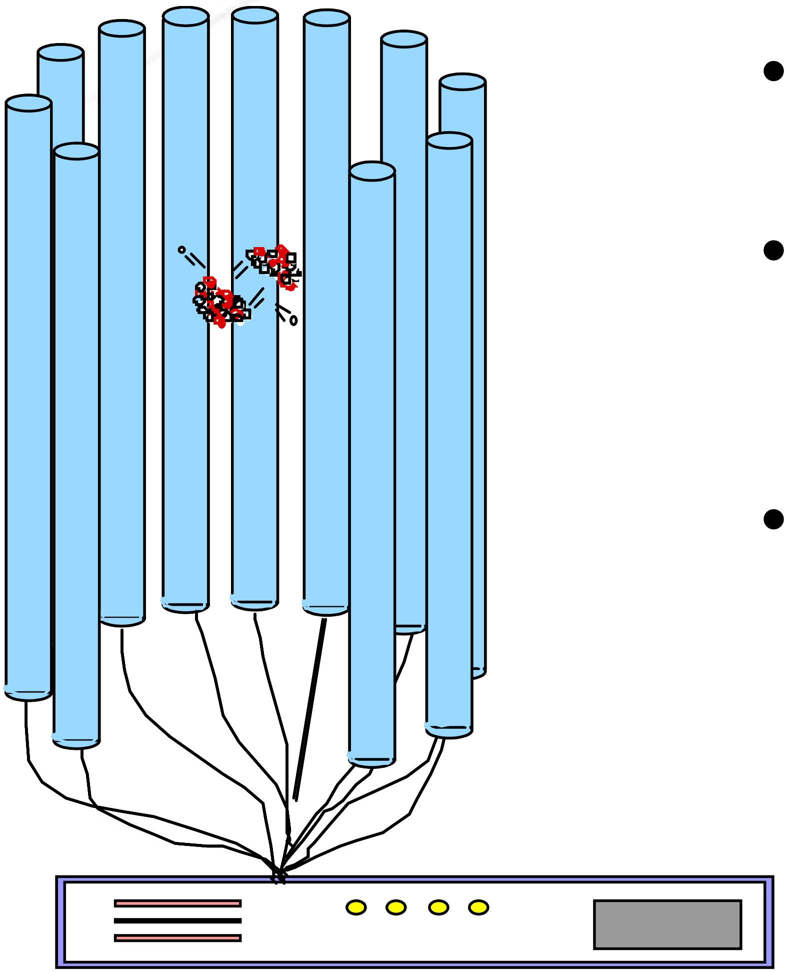
α - Ratio of (α,n) neutrons to SF neutrons

M_L - Leakage Multiplication

It is becoming clear that if we do not know any of theses values, we will three observables to uniquely determine each value and get a valid mass value.

Neutron Detector Systems – Theory & Concepts

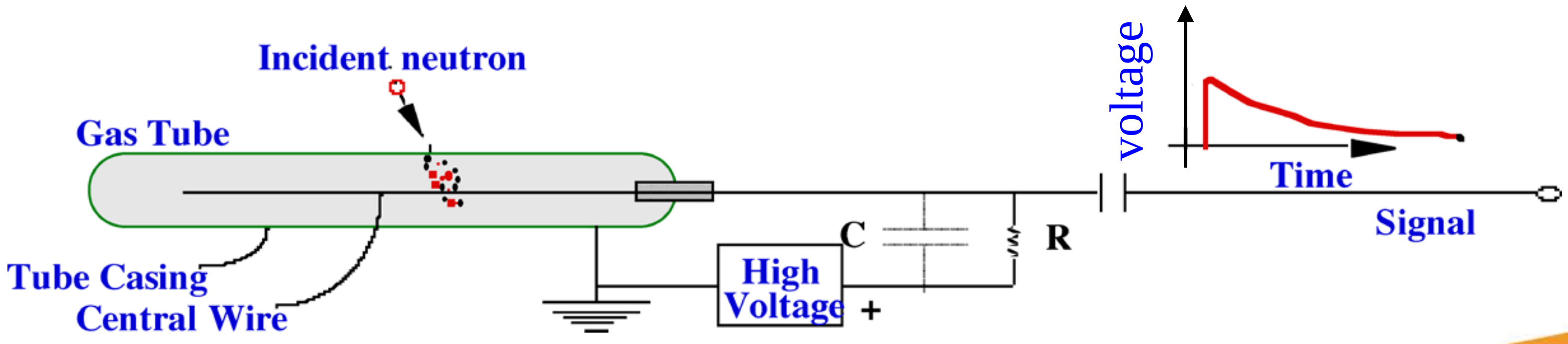
Neutron Coincidence Counter



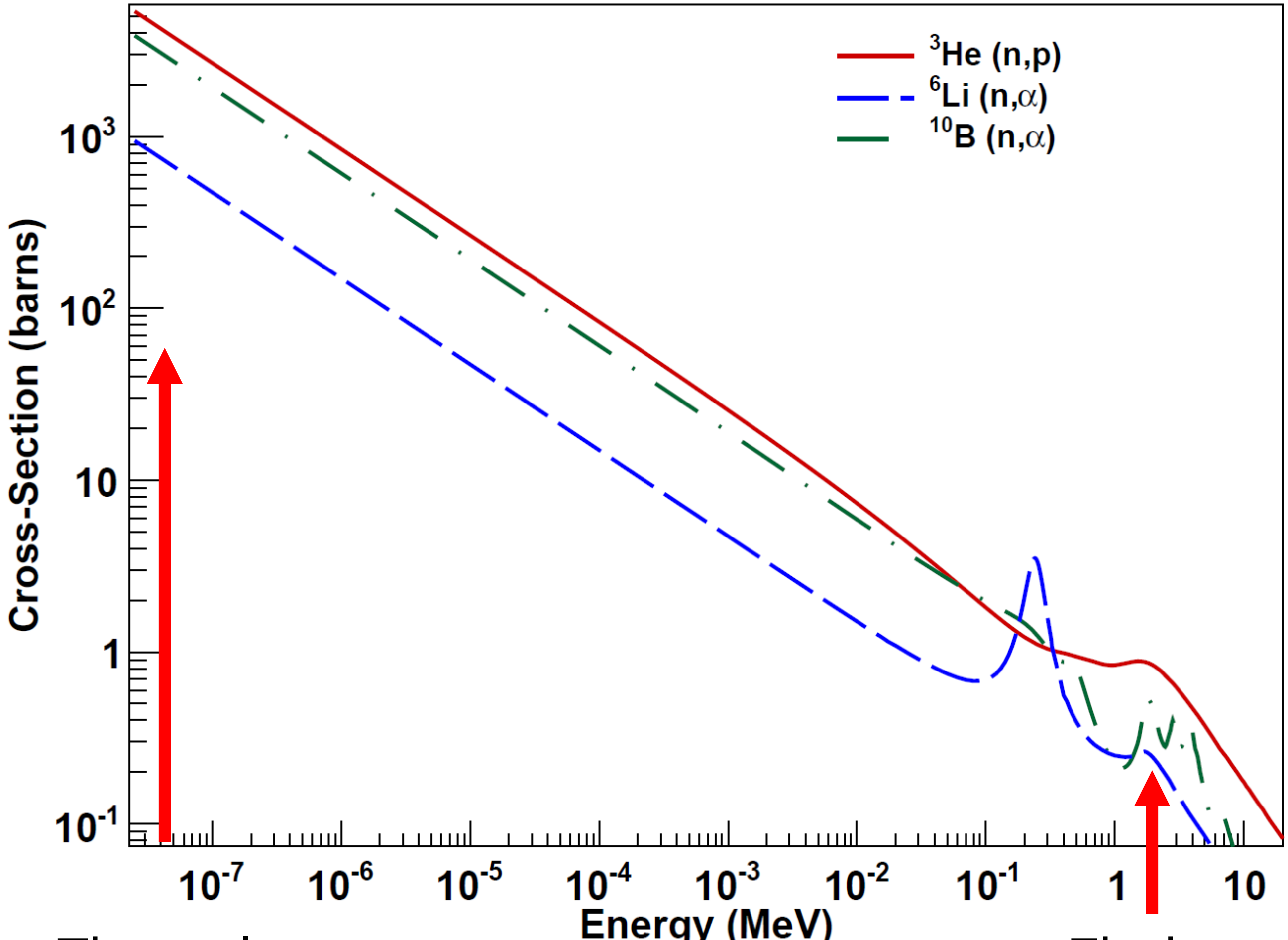
- Pu source surrounded by ^{3}He detectors
- Multiple neutrons emitted and detected as coincident neutrons
- Shift register counts coincident rate which is proportional to the $^{240}Pu_{eff}$ mass.

Neutron Detection

- Uses ^{3}He tubes imbedded in moderating material.
- Reaction is: $n + {}^3He \rightarrow p + {}^3H + 765$ keV
- Releases charge which is collected by gas tube.
- Detectors produce a distribution of electrical pulses.
- Electronics amplifies the pulses, sets threshold, and converts pulses above threshold to digital pulses.



Cross Section for Neutron Detection



Thermal

Fission

Moderation

- Moderation is a process by which a neutron collides with matter and loses energy. (from 2 MeV to 0.025 eV)
- The probability of neutron detection in ^{3}He is largest when the neutrons have energies near **thermal**.
- Most energy lost (best moderation) per collision when a neutron collides with nuclei of similar mass.
  - Polyethylene (CH_2)
  - Water (H_2O)
- Moderation to thermal usually takes many collisions
  - ~27 for polyethylene
  - ~119 for carbon
  - ~2175 for uranium
- Moderation takes ~2-5 µs
- Once moderated, neutrons diffuse throughout the detector system

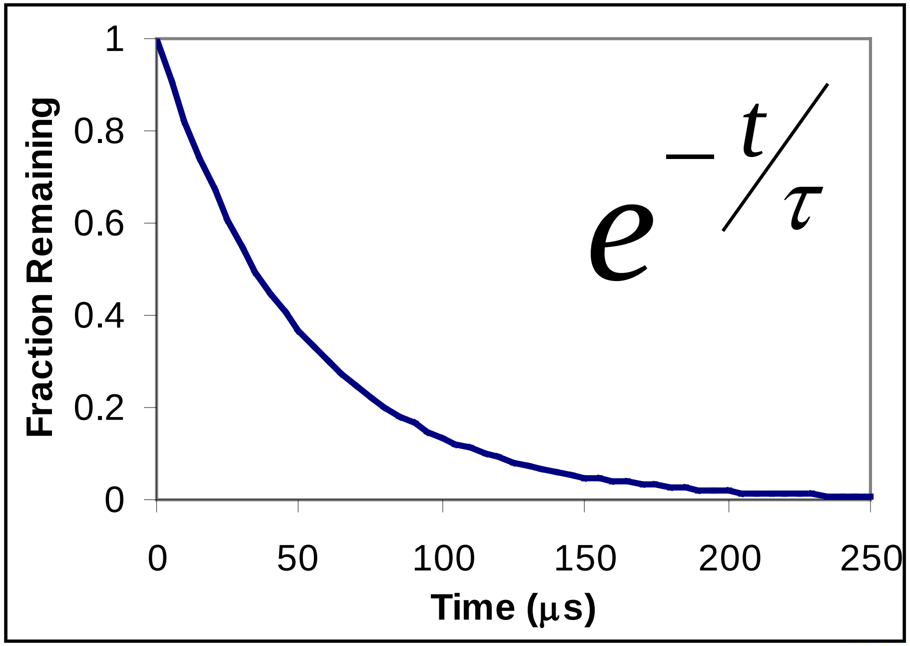
Die-Away Time

- After moderation, neutrons are lost in the detector by several processes:
  - Diffusing out of detector.
  - Diffusing to a ^{3}He detector tube and being absorbed.
  - Absorption by hydrogen or cadmium.
- Hydrogen both moderates and absorbs the neutrons.
- Time to travel 1 cm:
  - Fast neutrons (1 MeV) require 0.7 ns
  - Thermal neutrons (0.025 eV) require 4.5 μs (2200 m/s)

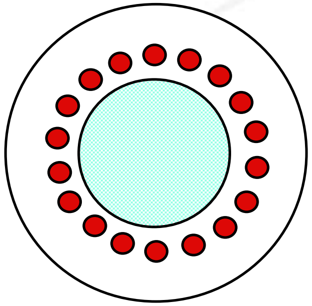
Die-Away Time

In most thermal detectors the neutron population decreases nearly exponentially in time. The time constant is called the **die-away time** **(τ)**.

For a detector with τ = 50 μs:



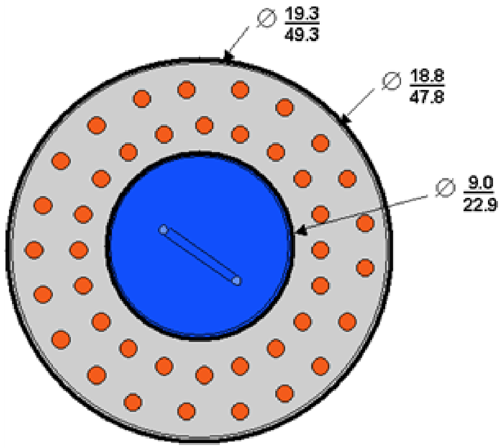
Detector Examples



HLNC

ε = 17.5%

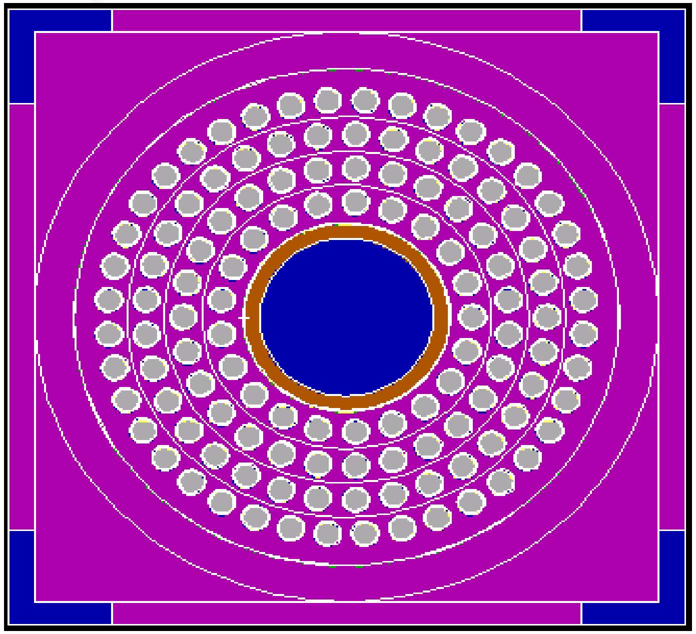
τ = 43 μs



AWCC

ε = 33%

τ = 51 μs

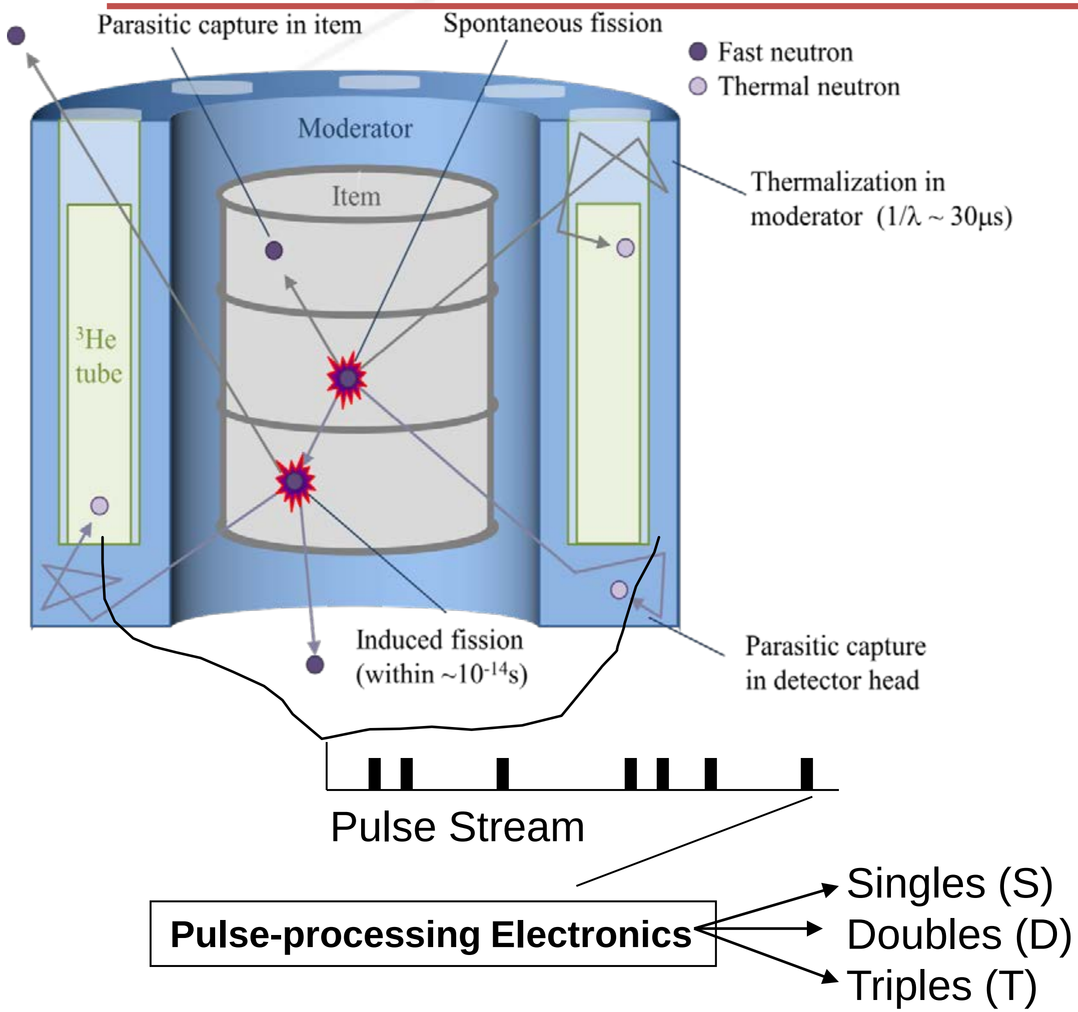


ENMC

ε = 65%

τ = 22 μs

Passive Neutron Counter



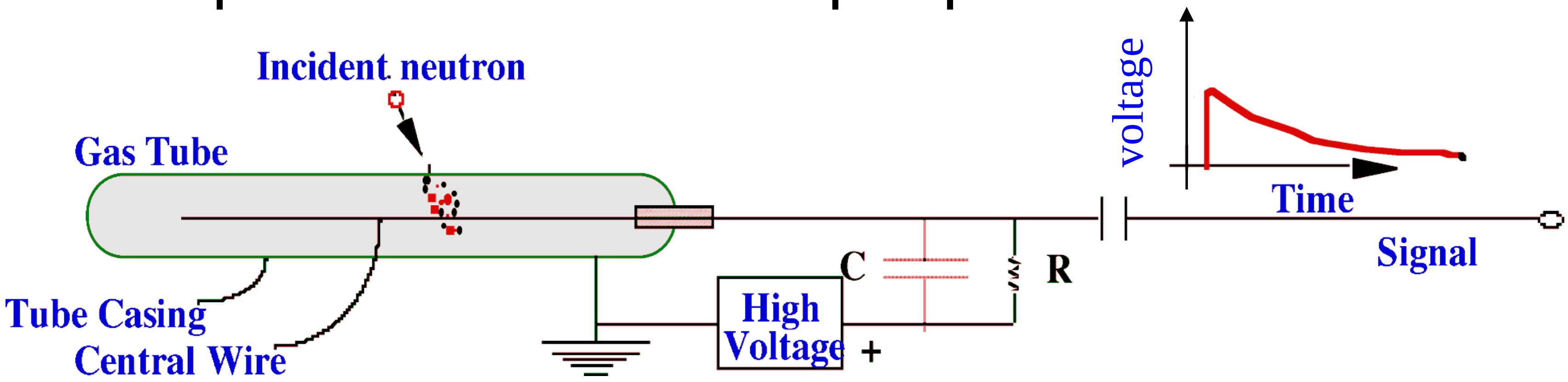
- Fissioning source surrounded by neutron detectors
- Neutron detectors are nominally ^{3}He proportional counters which have high detection efficiency for thermal neutrons
- To thermalize the neutrons, ^{3}He detectors are embedded in moderating matrix such as polyethylene.

Detector Systems – Some Important Details

System Deadtime

"Deadtime" results in the loss of detection information and can severely bias measurement results

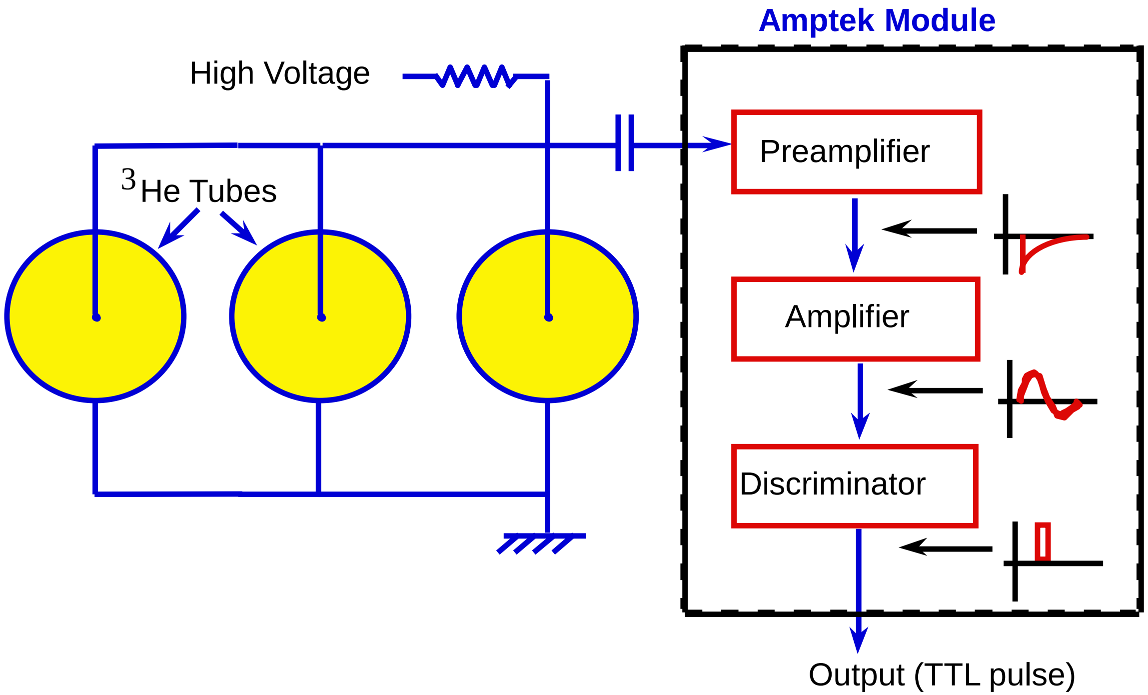
^{3}He tubes require 1-2 μs to recover before they can produce another output pulse.



System Deadtime

The Amptek amplifiers have an effective time constant of about 150ns.

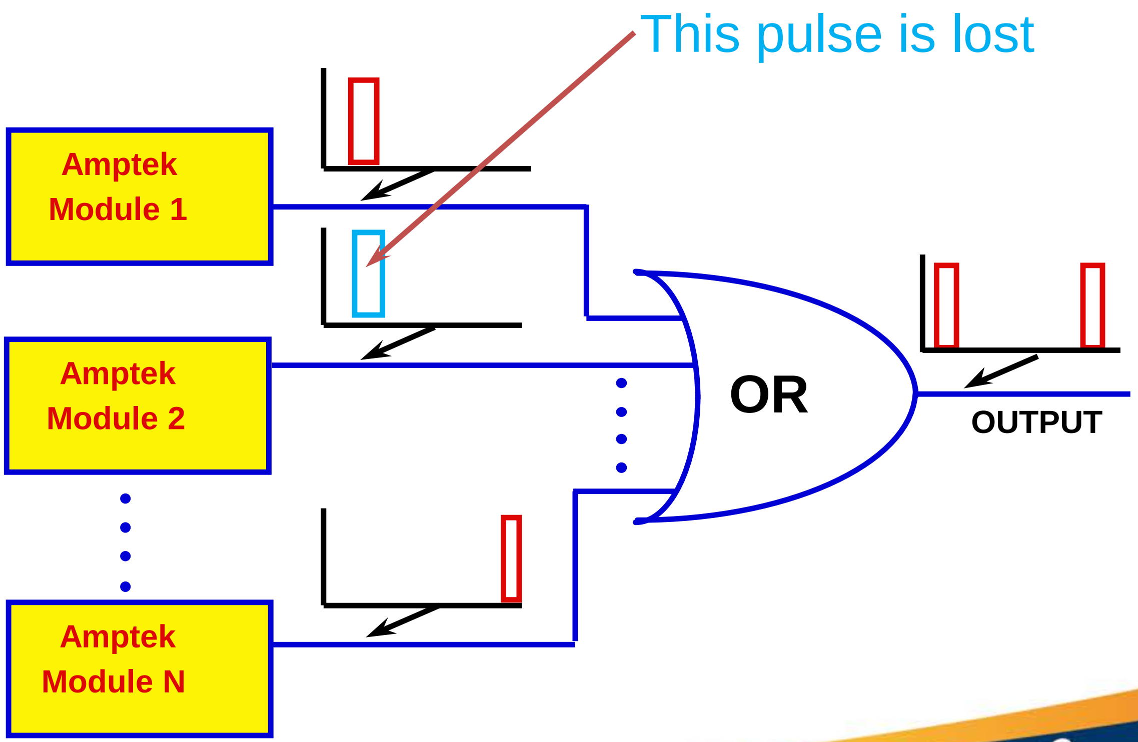
Deadtime losses can be minimized by using many amplifiers in parallel and varying the number of tubes per amp to balance the counting rates.



System Deadtime

If the 2-50ns wide TTL output pulses arrive at a summing OR gate within 50ns of each other, one will be lost.

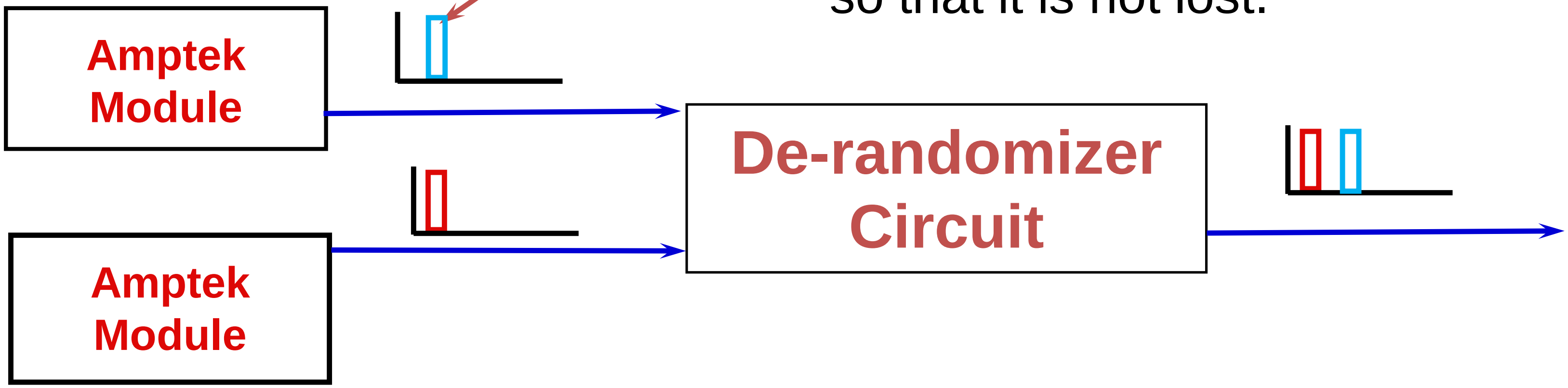
These losses can be minimized by replacing the OR gate with a circuit called a "de-randomizer."



Derandomizer Circuit

De-randomizer buffers hold pulses that are waiting to enter the shift register.

This pulse is delayed from entering the shift register circuit by 100ns, the clock speed of the coincidence gate, so that it is not lost.



The pulses are "quantized" into 100ns time bins. Their **"random"** appearance in time is removed or **"de-randomized."**

Deadtime Corrections

Even with elaborate measures to reduce deadtime, no detector system is free of deadtime losses. In standard coincidence counting, deadtime corrections are derived from careful characterization measurements of a series of calibrated reference sources. The corrections are of the form:

$$T_c = T_m e^{\delta T_m / 4}$$

$$R_c = R_m e^{\delta T_m}$$

where

$$\delta = A + BT_m$$

and the deadtime correction coefficients, A and B, are determined from the characterization measurements.

Multiplicity Deadtime

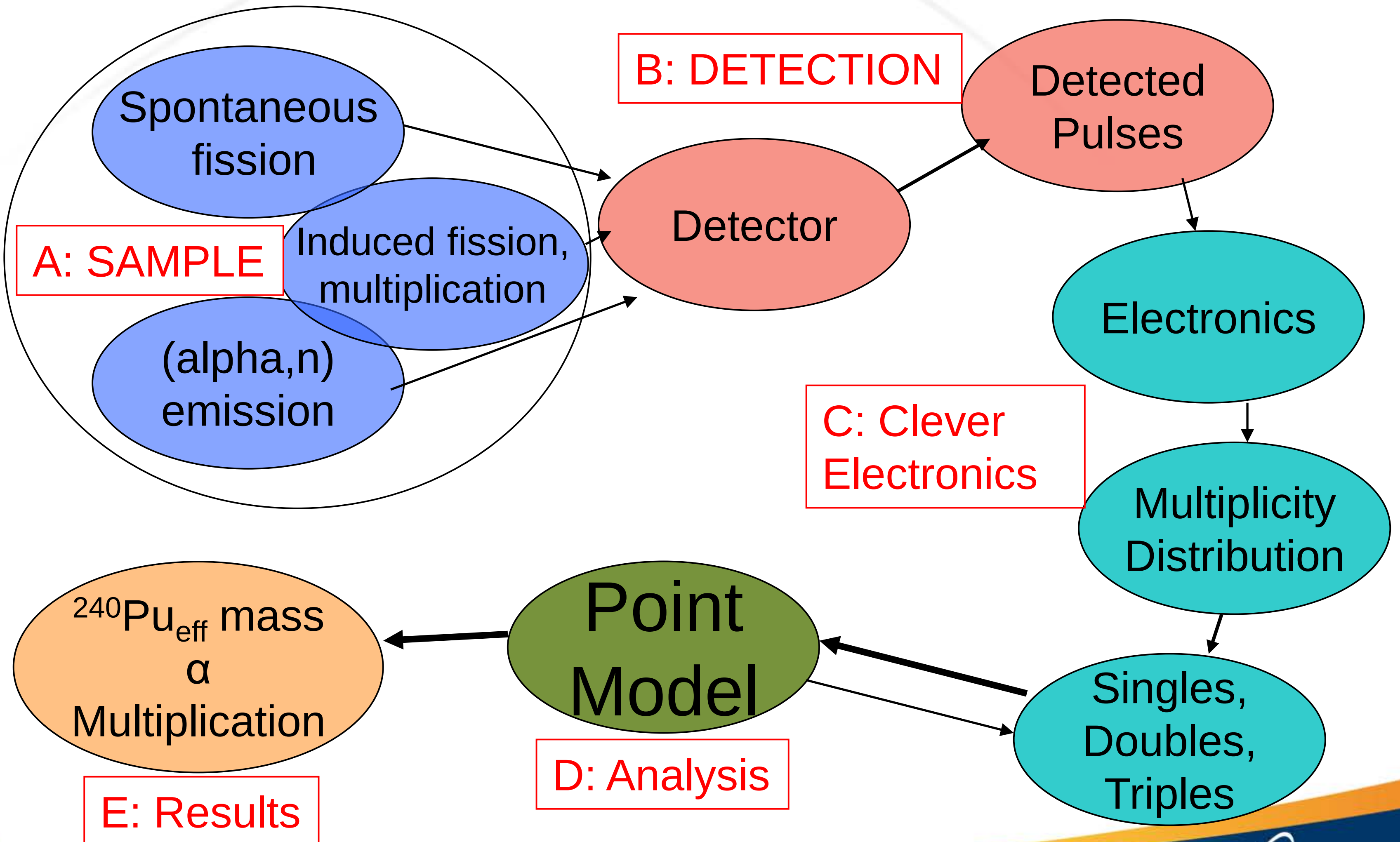
- The deadtime correction for triples counts is not as simple as our previous discussion on the topic - it depends on neutron correlations as well as the count rate.
- The current strategy is to "*minimize*" the problem by reducing the deadtime in the system by using many amplifiers and a de-randomizer circuit.
- A deadtime correction algorithm, developed by Dytlewski, is applied that depends on the effective "multiplicity deadtime" of the system.
- The multiplicity deadtime is determined using a series of ^{252}Cf sources.
- Multiplicity deadtimes in multiplicity counters that include modern de-randomizer circuits typically range from about 30 to 50 ns.

Multiplicity Counting Introduction

Multiplicity Counting Introduction

- Multiplicity Shift Registers use event-triggered gating to record the information in the pulse train (number of 0s, number of 1s, number of 2s, number of 3s, number of 4s, etc.) in a multiplicity histogram.
- We take the histogram and reduce it to Singles, Doubles and Triples.
- In order to interpret the results we use the "Point Model" to relate the item model-parameters ($^{240}Pu_{eff}$, α, M_L) and the detector model-parameters (ε, gate fractions) to the measured rates.

Multiplicity Counting Process

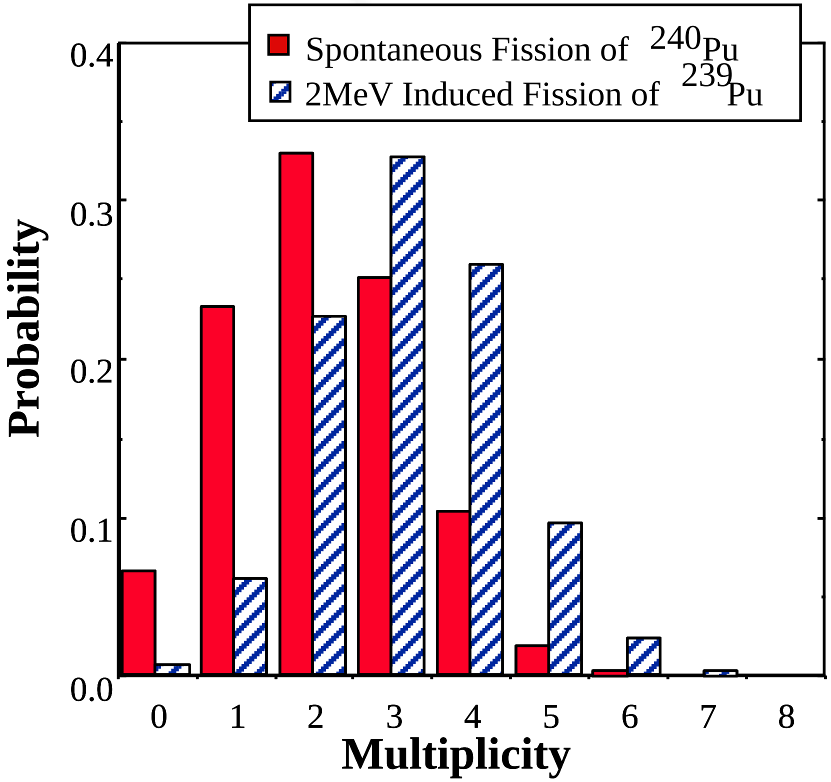


UNCLASSIFIED

Usefulness of Multiplicity Information

On average, Spontaneous Fission Neutrons are given off ~2 at a time.

On average, Induced Fission Neutrons are given off ~3 at a time.



The ratio of triples to doubles is a measure of multiplication because fission chains are created.

Utility of Multiplicity Information

But, even more importantly, *the distributions are different!*

In multiplicity counting we exploit this difference.

We measure the distribution of multiplicities in time.

Then we use what is known about these distributions and how the neutrons behave in the detector to statistically separate the three different types of neutron events.

Multiplicity Electronics and Example Distributions

Multiplicity Shift Register

A Multiplicity Shift Register is an extension of a coincidence shift register

- A multiplicity shift register works exactly the same as a normal coincidence shift register (Singles, Doubles)
- In Addition, for every trigger, the number of events present in the gates gets stored into histograms

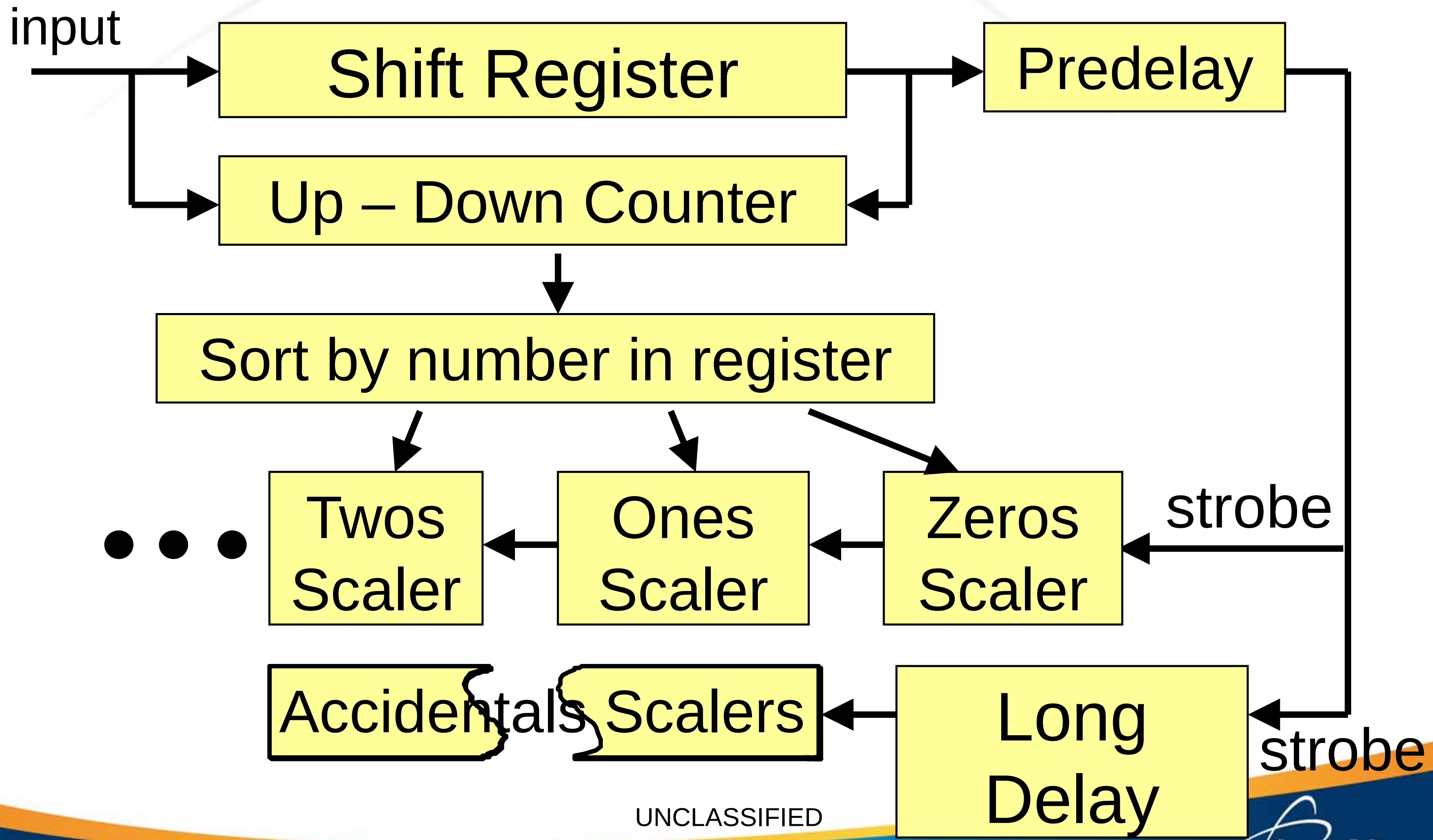
The histogram is a binning of the number of instances where bin-number of number of events were present in the gates (0, 1, 2, 3…512)

Multiplicity Shift Register

The Multiplicity Shift Register records two histograms

- One gate width immediately after the neutron is detected. This includes both real and accidental coincidences. Foreground Distribution.
- Second is after a long delay after the neutron is detected. This includes only accidental coincidences. Background Distribution.
- The measured S, D, and T count rates is determined from these two distributions.

Multiplicity Shift Register



Example Multiplicity Distribution

A 60g PuO_2 sample

Counter: FBLNMC

Time: 5000s

Gate: 32µs

Predelay: 3.0µs

Singles = 7431.1 cps

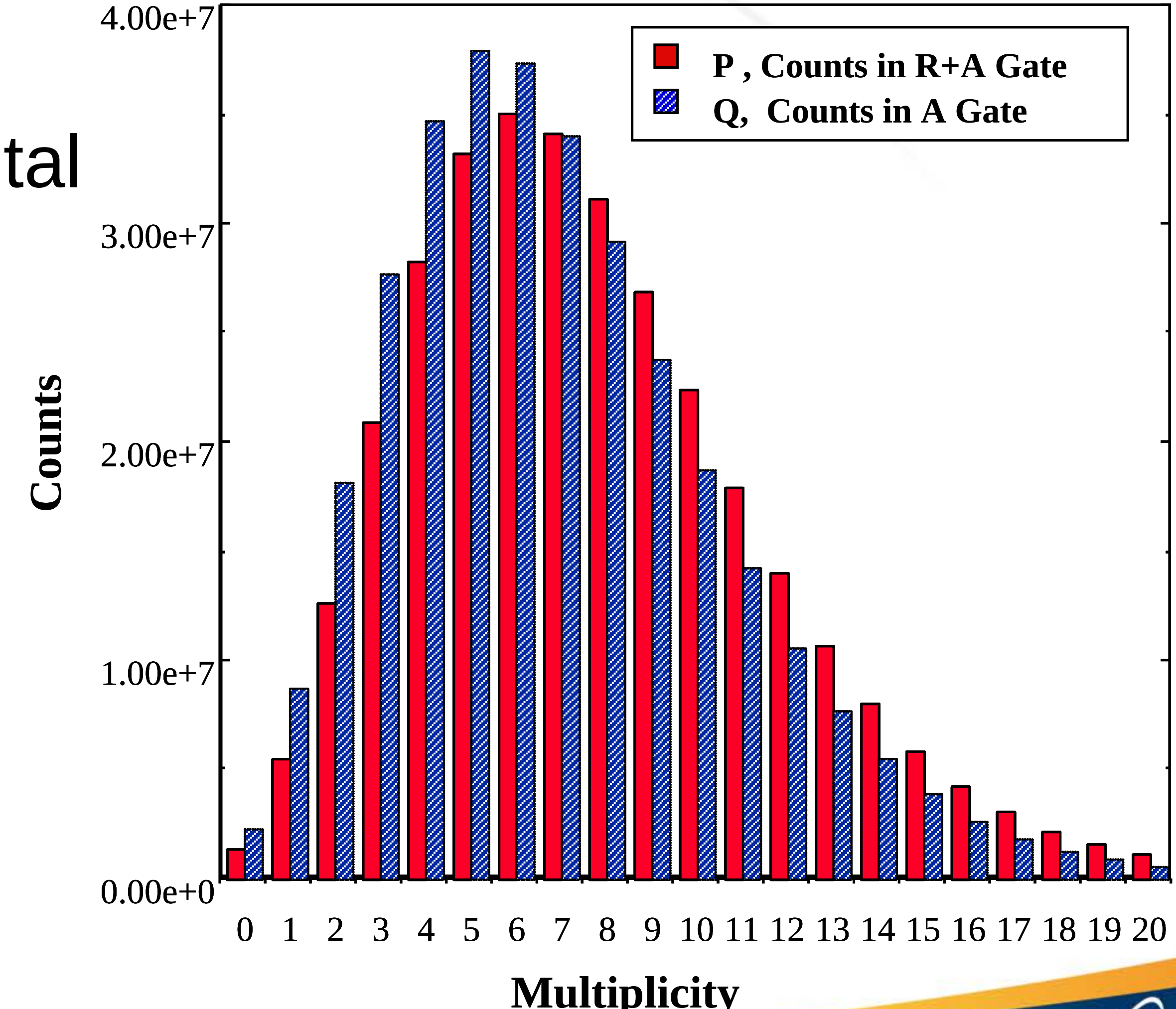
Doubles = 827.7 cps

Triples = 106.1 cps

	R+A	A
0	26804360	29731130
1	8187530	6222207
2	1772831	1016603
3	325270	157224
4	53449	22387
5	8231	3093
6	1237	402
7	183	42
8	30	8
9	2	1
10	0	0
11	0	0
12	0	0

Sample Multiplicity Distribution

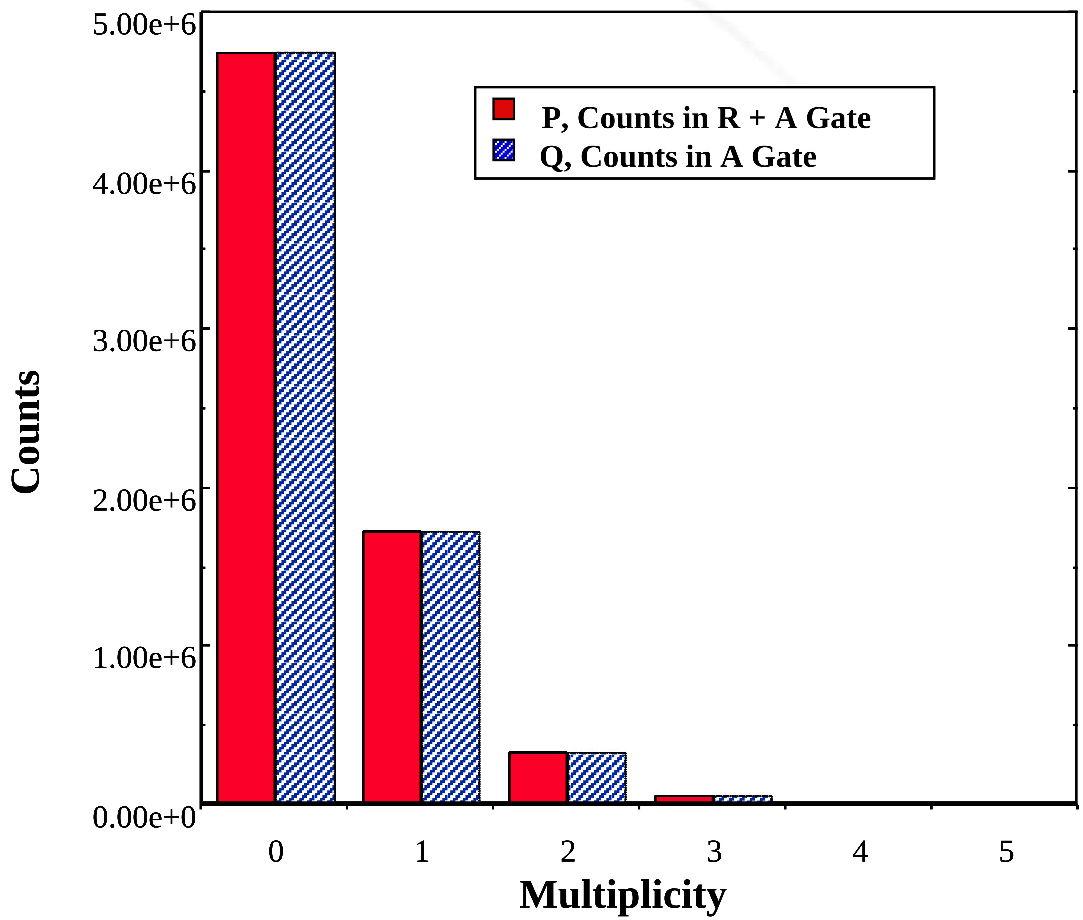
3.8 kg Pu metal



Sample Multiplicity Distribution

A (weak)
AmLi source

Notice that the foreground (R+A) and background (A) distributions are identical



Value of the Multiplicity Distribution

- These multiplicity distributions describe the probabilities of counting events of a given multiplicity in the R+A and A gates.

- The higher the singles rate, the longer the distributions will be. The average multiplicity of the accidentals distribution depends on this rate and the coincidence gate width.

- Real multiplicity information shifts the R+A distribution to higher multiplicities than the A distribution.

Singles, Doubles, and Triples from Measured Multiplicity Distributions

S, D, and T from the Distribution

- With the multiplicity shift register, we sample the multiplicity probability distribution of the measured neutrons.
- We have seen that there are two distributions in signal-triggered multiplicity measurement:
    - Foreground multiplicity distribution (R+A)
    - Background multiplicity distribution (A)

ν	foreground $P(\nu)$ R+A	background $Q(\nu)$ A
0	26804360	29731130
1	8187530	6222207
2	1772831	1016603
3	325270	157224
4	53449	22387
5	8231	3093
6	1237	402
7	183	42
8	30	8
9	2	1
10	0	0
11	0	0

Singles

- The singles counts (total number of neutrons collected) is simply ν_0 of the background distribution.
- “Background” distribution because in signal-triggered counting, the background records every trigger event.

$$\text{Measured S} = \sum_{\nu=0}^{\max} Q(\nu)$$

Doubles

- The doubles are the difference in the 1st moments of the multiplicity distributions in the R+A and A Gates.

$$Measured\ D = \sum_{\nu=1}^{\max} \nu P(\nu) - \sum_{\nu=1}^{\max} \nu Q(\nu)$$

- The doubles obtained this way are equivalent to the real coincidences obtained with a standard shift register circuit -- this provides a useful diagnostic for multiplicity shift register operation.

Triples

- The formula for calculating triples is intuitively much harder because the information in the R+A and A gates is correlated.
- The triples are the difference in the 2nd moments *minus* a cross-correlation term that depends on the doubles.

$$Measured\ \ T = \sum_{\nu=2}^{\max} \frac{\nu(\nu-1)}{2} \mathrm{P}(\nu) - \sum_{\nu=2}^{\max} \frac{\nu(\nu-1)}{2} \mathrm{Q}(\nu)$$

$$- \frac{\sum_{\nu=1}^{\max} \nu \mathrm{Q}(\nu)}{\sum_{\nu=0}^{\max} \mathrm{Q}(\nu)} \left(\sum_{\nu=1}^{\max} \nu \mathrm{P}(\nu) - \sum_{\nu=1}^{\max} \nu \mathrm{Q}(\nu) \right)$$

Example Calculation

	Measured Histograms		First Moments		Second Moments	
ν	R+A P	A Q	$\nu P(\nu)$	$\nu Q(\nu)$	$\nu(\nu-1)/2*P(\nu)$	$\nu(\nu-1)/2*Q(\nu)$
0	100	145	0	0		
1	200	208	200	208		
2	150	190	300	380	150	190
3	100	80	300	240	300	240
4	50	2	200	8	300	12
5	25	0	125	0	250	0
6	0	0	0	0	0	0
Sum=	625	625	1125	836	1000	442

Singles = 625 counts

Doubles = (1125 - 836) = 289 counts

Triples = (1000 – 442) – (836/625)(1125-836) = 171 counts

The Point Model Multiplicity Mathematics

Basic Passive Point Model Equations

$$S = Fm\varepsilon M\nu_{s1}(1+\alpha)$$

$$D = \frac{Fm\varepsilon^2 f_D M^2}{2}\left[\nu_{s2} + \left(\frac{M-1}{\nu_{i1}-1}\right)\nu_{s1}(1+\alpha)\nu_{i2}\right]$$

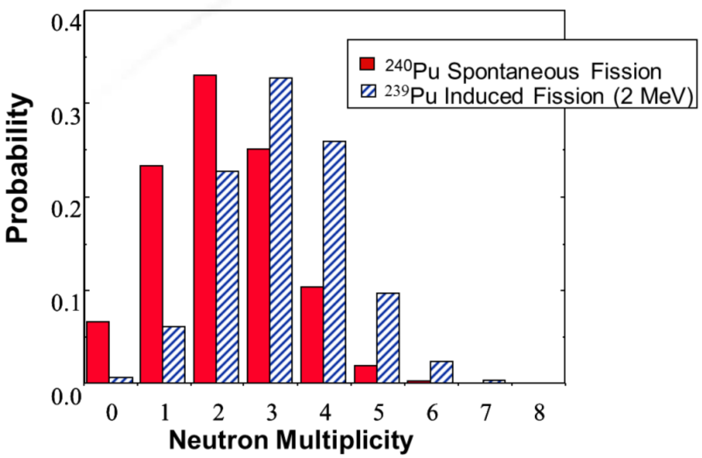
$$T = \frac{Fm\varepsilon^3 f_T M^3}{6}\left\{\nu_{s3} + \left(\frac{M-1}{\nu_{i1}-1}\right)\left[3\nu_{s2}\nu_{i2} + \nu_{s1}(1+\alpha)\nu_{i3}\right] + 3\left(\frac{M-1}{\nu_{i1}-1}\right)^2 \nu_{s1}(1+\alpha)\nu_{i2}^2\right\}$$

S = the observed singles rate in an ideal counter [neutrons/second],
D = the observed doubles rate [doubles/second],
T = the observed triples rate [triples/second],
F = the specific spontaneous fission rate of the sample [fissions/s-g]
= 473 fissions/s/g for ^{240}Pu,
ε = the neutron detection efficiency of the counter system [absolute units],
m = the mass of material [g]
M = The leakage multiplication of the sample [unitless],
α = the ratio of (α,n) neutron production to spontaneous fission neutron production in the sample [unitless],

f_D = the fraction of neutrons detected within the doubles gate time period [unitless],
f_T = the fraction of neutrons detected within the triples gate time period [unitless]
ν_{s1}, ν_{s2}, ν_{s3} = the first, second, and third reduced moments spontaneous fission neutron distribution,
ν_{i1}, ν_{i2}, ν_{i3} = the first, second, and third reduced moments induced fast fission neutron distribution

UNCLASSIFIED

Moments of Fission Distribution



The Moments for ^{240}Pu Spontaneous Fission

ν_{s1} = 2.154

ν_{s2} = 3.789

ν_{s3} = 5.211

The Moments for ^{239}Pu Induced Fission (2 MeV)

ν_{i1} = 3.163

ν_{i2} = 8.240

ν_{i3} = 17.321

These moments are calculated using the same moments analysis we used before.

The values are:

ν_{x1} - the average number of single neutrons per fission event,

ν_{x2} - 2! times the average number of neutron pairs per fission event, and

ν_{x3} - 3! times the average number of neutron triplets per fission event.

Calibration of a Multiplicity Counter

For a Cf source ($M = 1$, $\alpha = 0$) the multiplicity equations reduce to simple forms.

$$S = F\varepsilon\nu_{s1}$$

$$D = \frac{1}{2}Ff_d\varepsilon^2\nu_{s2}$$

$$T = \frac{1}{6}Ff_t\varepsilon^3\nu_{s3}$$

With F known (calibrated source), can solve for the unknown detector characteristics, ε, f_d, and f_t.
Where:

$\nu_{s1} = 3.75$
$\nu_{s2} = 11.96$
$\nu_{s3} = 31.81$

In principle, standards are not needed to calibrate a multiplicity counter

Multiplicity Counting Analysis

- We are now armed with:

  Three Observables: S, D, and T
  Three Unknowns: $^{240}Pu_{eff}$, α, M_L

- We know how to extract the observables from the measured data

- We have the mathematical model relating the observables to the unknowns.

- The rest is just algebra…..

Reducing the Point Model Equations

Solve the point model equations for *M*:

$$a + bM + cM^2 + M^3 = 0$$

where:

$$a = \frac{-6T\nu_{s2}(\nu_{i1} - 1)}{\varepsilon^2 f_t S(\nu_{s2}\nu_{i3} - \nu_{s3}\nu_{i2})}$$

$$b = \frac{2D[\nu_{s3}(\nu_{i1} - 1) - 3\nu_{s2}\nu_{i2}]}{\varepsilon f_d S(\nu_{s2}\nu_{i3} - \nu_{s3}\nu_{i2})}$$

$$c = \frac{6D\nu_{s2}\nu_{i2}}{\varepsilon f_d S(\nu_{s2}\nu_{i3} - \nu_{s3}\nu_{i2})} - 1$$

See?
Algebra

Solving for Mass and Alpha

Once M is found, solve for the mass by:

$$Fm = \frac{\left[\frac{2D}{\varepsilon f_d} - \frac{M(M-1)\nu_{i2}S}{\nu_{i1}-1}\right]}{\varepsilon M^2 \nu_{s2}}$$

And for alpha by:

$$\alpha = \frac{S}{\varepsilon F m M \nu_{s1}} - 1$$

Extra Slides